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19. ABSTRACT (Continue on reverse if necessary and identify by block number) <p>The objective of this study has been to identify the emission mechanism in light emitting tunnel junctions by comparing experimental results with models of the mechanism. Work has concentrated on Au-oxide-Al junctions almost exclusively because of their durability and the richness of their spectra. The emission spectrum has been measured as a function of bias voltage for about one hundred junctions.</p> <p>It was necessary to determine if any of the three surface-plasmon-polariton (SPP) modes a junction supports is primarily responsible for the emitted light.</p> <p>Our experiments show that the dominant mode is the slow mode that has maximum field strength in the barrier. The Au fast mode and the Al fast mode, with maximum field strengths at the Au-air interface and Al-substrate interface are only (continued on back)</p>			
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Final Report on Study of Light Emitting Tunnel Junctions

ARO Proposal #23493-PH

Contract #DAAL03-86-K-D084

At this stage, the experimental work on this project is complete and the data analysis is nearly complete.

Technical Results

The objective of this study has been to identify the emission mechanism in light emitting tunnel junctions by comparing experimental results with models of the mechanism. We have concentrated on Au-oxide-Al junctions almost exclusively because of their durability and the richness of their spectra. We have measured the emission spectrum as a function of bias voltage for about one hundred junctions with film thicknesses given in Table I. It was necessary to look at a large number of samples because of the rather large variations in the emission from sample to nominally identical sample, see Figure 1.

There are two central questions in the light emission process. First, which if any of the three surface-plasmon-polariton (SPP) modes a junction supports is primarily responsible for the emitted light? Second, how do the tunneling electrons excite the SPP modes?

Our experiments show that the dominant mode is the slow mode that has maximum field strength in the barrier. The Au fast mode and the Al fast mode, with maximum field strengths at the Au-air interface and Al-substrate interface are only marginally

involved. These conclusions are independent of any attempt to calculate the emission. I am just now completing an attempt to calculate the emission spectrum in a way that identifies the processes that dominate the emission. Although this is not yet finished, results to date support the conclusion that the slow mode is mainly responsible. They suggest that the scattering of the slow mode electric field parallel to the plane of the junction off roughness at the edges of the barrier is the primary coupling mechanism for the mode. Furthermore, I think that when the modelling is complete, it will show that only inelastic tunneling current fluctuations in the barrier will be needed to explain the emitted light. Inelastic processes in the metal films probably will not be needed to make the model conform to the data.

The identification of the slow mode proceeded as follows. First, the measured spectra off the Au side of the junction are nearly independent of the thickness of the Al film. This eliminates the possibility of the Al fast mode being a substantial contributor to the Au side emission. The converse is also true. The spectrum out the aluminum side of the junction is nearly independent of the thickness of the Au film. This eliminates the Au fast mode as a major contributor to the Al-side emission. The remaining possibility for emission from the Au surface is that it is due either to the Au fast mode or the slow mode or both. To sort this out, we have measured the emission spectra from junctions with the Au film covered with 10 nm of amorphous Ge. Attenuated-total-reflection (ATR) measurements on

these samples show no evidence of a Au fast mode SPP. The absorption dip and the resonant enhancement of the transmitted light due to scattering of the SPP mode off residual surface roughness are both less than 1% of their values on junctions without the a-Ge layer. We take this as proof that the 10 nm a-Ge layer has destroyed the Au-fast mode SPP. Despite the elimination of that mode, the emitted intensity and the energy dependence of the spectrum on a-Ge covered and bare junctions are nearly identical. See Figure 2. This shows that the light radiated from the Au side of the junction is due entirely to the slow mode. It also hints that slow mode scattering of the roughness on the surface is not important in the emission mechanism. The Al fast mode is not so easily extinguished, because of the very large negative value of ϵ_1 for Al. The following circumstantial evidence indicates that the Al fast mode plays at most a small part in the emission process of the Al surface. The spectra, like those in Figure 1, suggest two superposed peaks. Both peaks are present in the spectra measured off both sides of the junction. However, the ratio of the Al side spectrum to the Au side spectrum is a monotonic function of energy and the sample to sample variations are far smaller than are the variations in the spectra themselves. See Figure 3. The cancellation of the peaks in the ratio suggests that the spectra off both sides have a common origin. Since the slow mode is the source of the light off the Au side, it must also be the source off the Al side. Except for the implicit acceptance of the general features of the fields in each SPP mode, the above

discussion is independent of any theoretical models.

Attempts to model the junctions are nearly complete. Clearly one would like to identify some feature of the slow mode that contains at least one peak near 2.0V. Figure 4 shows the square of the component of the slow mode field parallel to the planes of the film at the Au-oxide interface. It is calculated assuming that a drive current of unit strength and proportional to $\hat{z} e^{ik_{\parallel} x} e^{-i\omega t}$ exists in the barrier, where \hat{z} is normal to the junction. The figure shows the square of the parallel field as a function of k_{\parallel} and ω . The ridge defines the dispersion curve of the slow mode and the peak in the response (high point on the ridge) is at 1.98V. I am attempting to calculate the scattering of the slow mode field off roughness at the barrier to compare to the data. But even the result of Figure 4 encourages the point of view that the slow mode is responsible for the emission. I will try to find both peaks in the model and check to see if the model yields values of the Al side to Au side ratio that imitate the data before the work outlined above is submitted for publication. The work will be completed shortly.

The experiments described above have given us ample opportunity to assess the light emitting junctions as practical devices. Some of our samples have been quite bright, so there is hope that the lack of intensity can be overcome. A more worrisome problem are aging effects. Our best junctions could be run for two days. Our typical junctions would run only several hours. As a function of time two things happened. First the resistance of the junction increased so that at constant bias

voltage the current steadily decreased. Moreover, the efficiency defined as the number of photons emitted per tunneling electron also decreased. Both effects occurred regardless of whether the oxide was formed thermally or by plasma oxidation. Both occurred regardless of whether the junctions were run in room air, in vacuum, or in liquid ^4He . Charged species in the barrier must be moving in response to the large DC electric field the bias voltage creates. If the species are intrinsic rather than impurities, using light emitting junctions as light sources may be impractical. We were unable to pursue this issue further.

In the course of our work we pursued two side lines. First, we used a scanning tunneling microscope to measure the surface roughness on evaporated films and junctions. We believe we were the first to use an STM for this purpose. The results were reported earlier. Here I will comment that if it turns out it is roughness at the barrier that counts, as the experiment with the a-Ge suggests, the STM will not help much in studying light emitting junctions. Secondly, we have done an extensive set of measurements of ATR response of Au films. In the course of the modelling, it became apparent that at 632.8 nm, the dispersion relation for the Au fast mode has no root for films thinner than 43.8 nm. Such films still exhibit ATR dips and enhanced transmission at the ATR angle. If SPP's are the roots of the dispersion relation, the question is how are the ATR data associated with the properties of the SPP? The analysis of this data will be finished after the junction modelling described above is complete.

One of the goals of the proposal was to make precision measurements of the derivatives of the I-V curves to help identify the inelastic tunneling mechanism. The attempts to do this were completely frustrated by the aging effects discussed earlier.

Scientific Personnel

The contract supported Dr. Patricia D. Sparks as a postdoctoral researcher. She is now an Assistant Professor at Harvey Mudd College.

It also supported Mr. Derrick Russel, a beginning graduate student, as a Graduate Research Assistant.

Publications

The following publications have appeared.

- 1) Two-mode Radiation from Light Emitting Tunnel Junctions, R. M. Pierce, J. E. Rutledge, and S. Ushioda, Phys. Rev. B36, 1803 (1987).
- 2) Visual Observation of a Voltage Distribution, P. D. Sparks and R. M. Pierce, Am. J. Phys. 56, 513 (1988).
- 3) Frequency Cutoff of Emitted Light from Tunnel Junctions, P. D. Sparks and J. E. Rutledge, Bull. Am. Phys. Soc. 32, 872 (1987).
- 4) Surface Plasmon Polaritons in Light Emitting Metal-insulator-metal Tunnel Junctions, P. D. Sparks and J. E. Rutledge, Bull. Am. Phys. Soc. 33, 224 (1988).

In preparation are

- 1) A paper describing the tunnel junction work outlined in this report. It is tentatively titled "Experimental Determination

of the Dominant Mode in Light Emitting Tunnel Junctions." It will be authored by P. D. Sparks and J. E. Rutledge and submitted to Physical Review B.

- 2) A paper on the ATR of the Au films. It does not yet have a title. The author will be J. E. Rutledge and it will be submitted to Physical Review B.
- 3) A paper on the behavior of anomalous tunnel junctions. The authors will be P. D. Sparks and J. E. Rutledge. It will be submitted to Applied Optics.

Preprints and reprints of these will be sent to ARO, unless we are requested not to do so.

Table I

Al Thickness →		90	180	380	750	1300
Au Thickness	↓					
90		X				
180			X	X		
300			X	X	X	X
770				X	X	
1300			X	X		

Figure Captions

- Figure 1. The emitted spectra from two nominally identical 30 nm Au and 380 nm Al tunnel junctions. Note the difference in scales. Both junctions are biased at 2.8V. Both spectra consist of two peaks, one centered near 1.9V and the other near 2.1.
- Figure 2. The spectrum emitted from the Au side of a 30 nm Au and 130 nm Al junction before and after 10 nm of a-Ge are deposited. Their intensities are identical within the usual sample to sample scatter and the energy dependence is nearly identical.
- Figure 3. The emitted spectra off the Al side divided by the emitted spectra off the Au side for the junctions of Figure 1. Notice that the peaks are gone and the ratios differ by 50% whereas the spectra differ by a factor of 6.
- Figure 4. A contour plot of the square of the component of the electric field parallel to the junction's surfaces. The current driving the mode is assumed to be $\frac{1}{2} \hat{z} e^{ik_{\parallel} x - iE/\hbar t}$, confined to the barrier. The calculation is for a Au thickness of 30 nm and an Al thickness of 38 nm. The barrier is assumed to be 3 nm thick. The peak is at 1.98 eV and may be the peak seen in the emission spectrum.

Fig 1

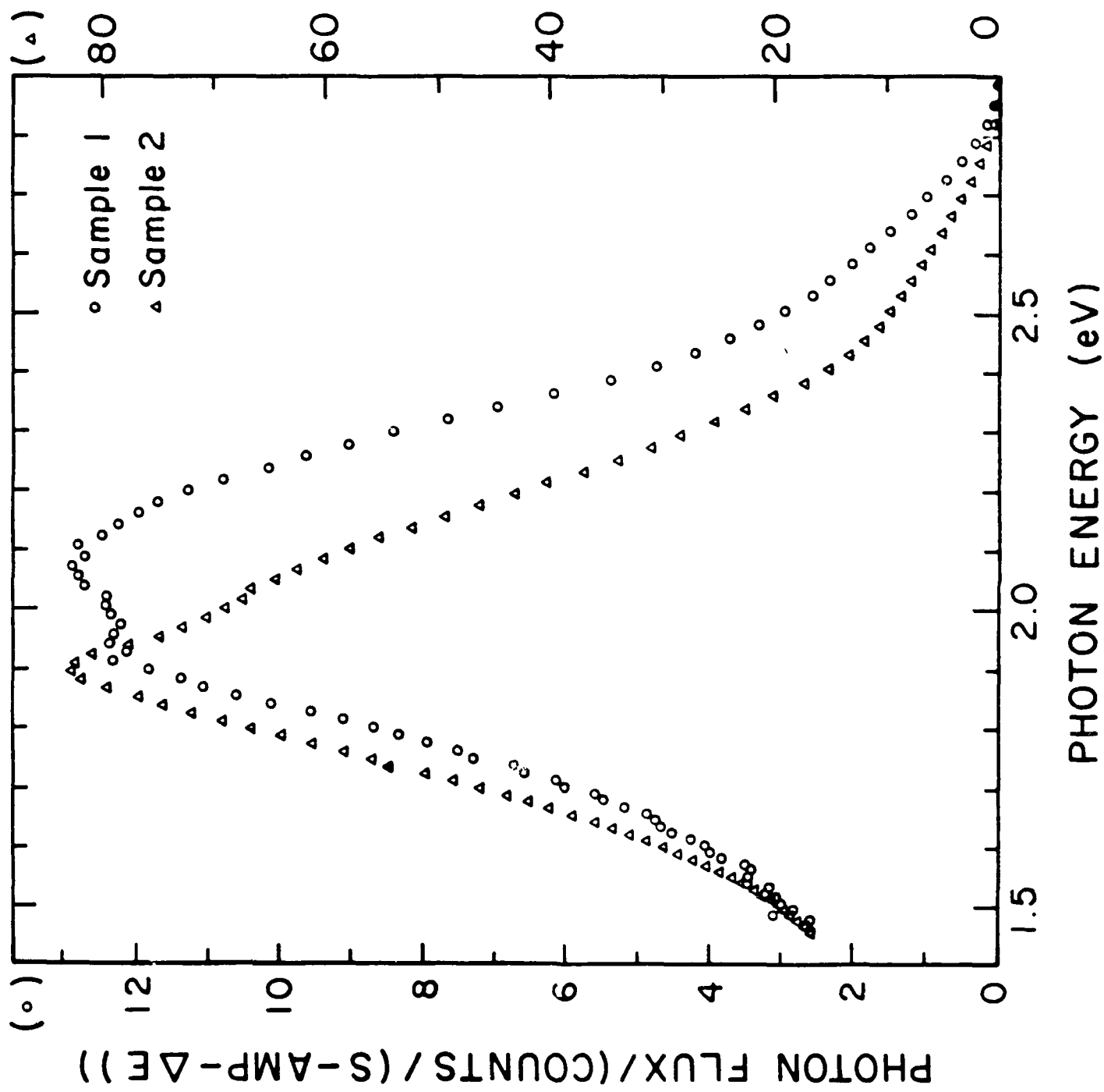


Fig 2

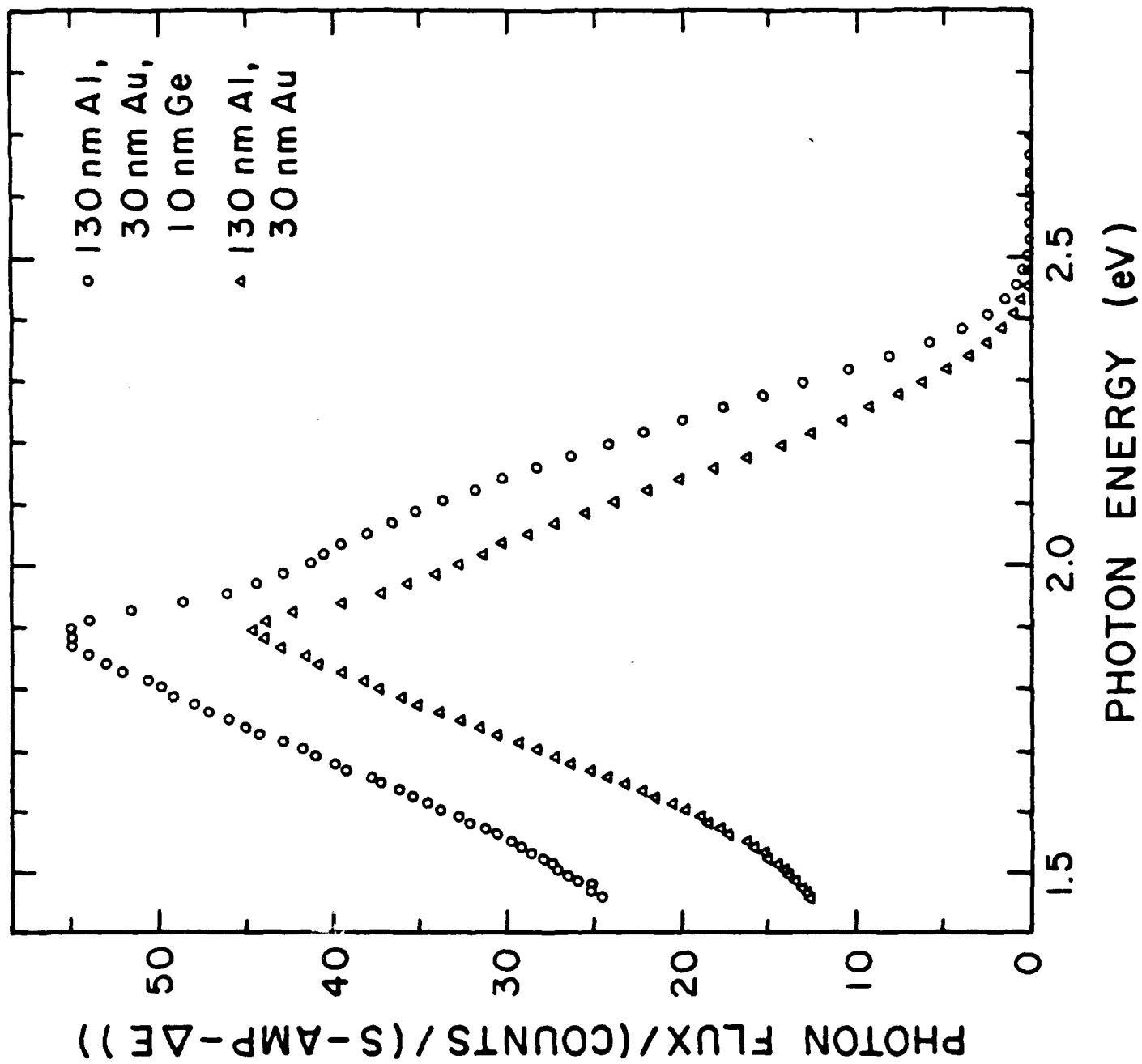


Fig 3

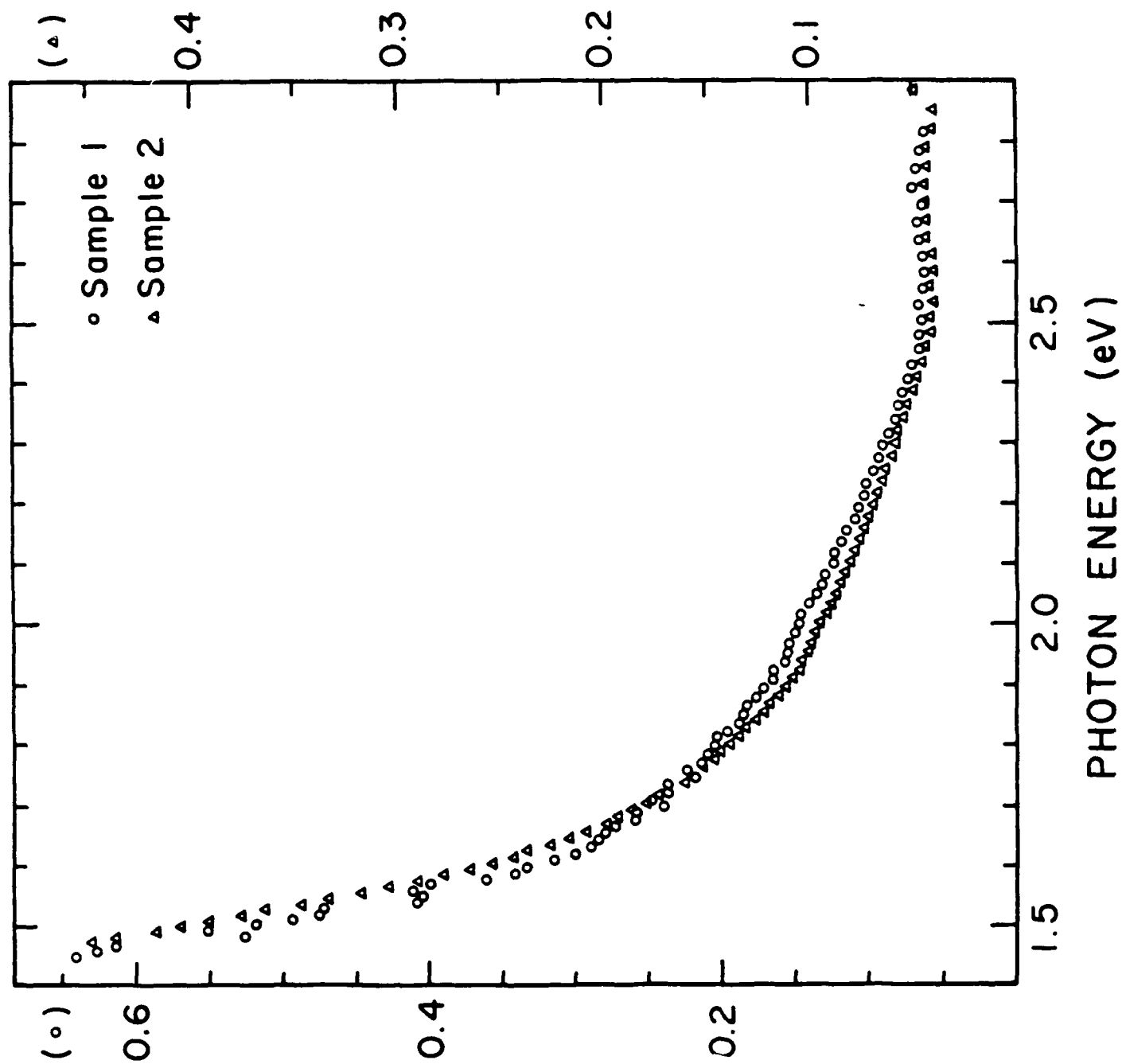


Fig 4

